Treatment of NOx from Diesel Engine Exhaust by Dielectric Barrier Discharge Method

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***Abstract*— This paper reports improved performance of discharge plasma in filtered engine exhaust treatment. Our paper deals about the removal of NO_x emissions from the diesel exhaust by electric discharge plasma. For the treatment of diesel exhaust a new type of reactor referred to as cross-flow dielectric barrier discharge reactor has been used, where the gas flow is perpendicular to the corona electrode. Experiments were conducted at different flow rates ranging from 2 l/min to 10 l/ min. The discharge plasma assisted barrier discharge reactor has shown promising results in NO_x removal at high flow rates.**

***Index Terms*— Electric Discharge, Dielectric Barrier Discharge Cross Flow Reactor (CFR), NO_x removal, Filtered diesel exhaust.**

I. Introduction

Diesel Engines are used widely nowadays for heavy duty transportations and many more off-road applications owing to their low operating costs, good thermal efficiency and long durability. However, they emit NO_x, CO, CO_2 and few other gases which are harmful and have led to widespread pollution. Diesel engines alone account for nearly 48 percent of India's fuel consumption, henceforth contributing significantly to the man-made air pollution. One of the most harmful pollutants present in the diesel exhaust is NO_x. The conventional techniques which are available to control emission now are either difficult to operate or does not satisfy the stringent emission standards. Reducing diesel engine exhaust to meet future emission standards is a challenging task and there is need for better after-treatment techniques [1]. For controlling NO_x, the electric discharge based plasma is an upcoming technology being used mainly. Non-thermal plasma technologies can be grouped as surface discharge, pulsed streamer corona discharge (PSC), dielectric barrier discharge (DBD) [2, 3], and packed-bed corona discharge (PCP). These techniques have shown promising results owing to their rapid reactions, high electron energies and easy operation. Y S Mok et al conducted the experiment in presence of O_3 in the DBD reactor which when mixed with NO_x is more energy efficient [4]. M. Okubo et al used a barrier type packed bed non thermal plasma application driven by a pulse high voltage power supply under oxidation for simultaneous removal of NO_x and PM [5].

The objective of our work has been to investigate on the suitability of the electric discharge technique for the engine exhaust treatment in actual diesel engine running conditions using a dielectric barrier discharge plasma. The electric discharge or corona plasma method has lots of advantages when compared to its counterpart as it is cost effective, low capital and operation costs, salable by-products, and integration with the existing systems. In this paper we describe an alternate reactor geometry referred to as cross-flow DBD reactor, where the exhaust gas flow perpendicular to the wire-cylinder reaction chamber. This reactor is used to treat the actual exhaust of a 3.75 kW diesel-generator set. The main emphasis is laid on the NO_x treatment of diesel engine exhaust. Experiments were carried out at room temperature at different gas flow rates at no load.

II. Experimental Setup

The schematic of experimental setup used during the present research work is shown in Fig. 1. Studies were carried out at the laboratory scale using a diesel generator set as the source of the exhaust and main emphasis has been given on removal of NO_x. Sample gas from the diesel engine exhaust pipe is first allowed to pass through steel wool where the coarse oil mist and dust particulate gets trapped. The sample is then allowed to flow through a conditioning system for filtration of residual water droplets present in the exhaust. The gas is then made to pass through a particulate filter to trap solid particles up to the size of 5 ìm. The filtered gas is then treated in various ways and sent to the gas analyzer for analysis of its concentration.

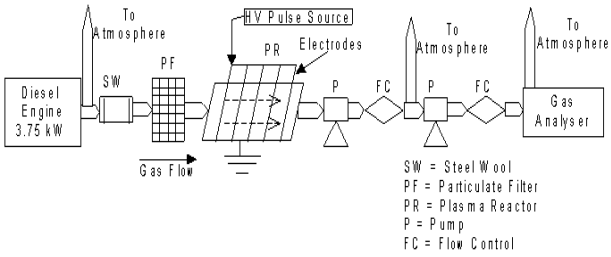

Figure 1. Schematic of experimental setup used for the treatment of exhaust by discharge plasma.

A. Diesel Engine

The diesel exhaust for the treatment was taken from a 5HP, 3.75kW diesel generator. A single phase alternator of 2.2kVA fed to lighting load. The diesel engine is treated at no load. Most of the exhaust is let off to atmosphere and only a part of the exhaust is taken for the analysis. Here the flow rate is maintained up to 10 l/min for the analysis part of the experiment.

B. Filtering System

The filtering system consists of:

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- Steel Wool
- Particulate filter

Steel Wool is used to remove oil mist and dust particulate off the exhaust. Particulate filter removes solid particles of the size of 5 micron from the exhaust gas.

C. High Voltage Pulse Source

The power source consist 230V AC input that is stepped up using a step-up transformer and then converted to DC using a diode rectifier unit. A charging capacitor is used to smooth out the output DC. The high voltage DC output is fed to RSG that produces pulses of rise time 20 ns and 75 pulses per second (pps). The pulse generating circuit is shown in Fig. 2. When the rotating electrode of the RSG is aligned with the fixed line electrode, the charge on c_g is discharged to plasma reactor. The hemispherical rotating electrode of the RSG was connected to a motor through an insulating rod. By changing the speed of the motor, the frequency of the pulses applied to the plasma reactor can be controlled. The voltage-time diagram has been shown in Fig. 3.

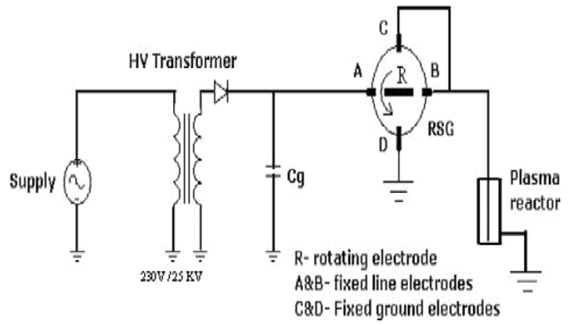

Figure 2. Pulse generating circuit

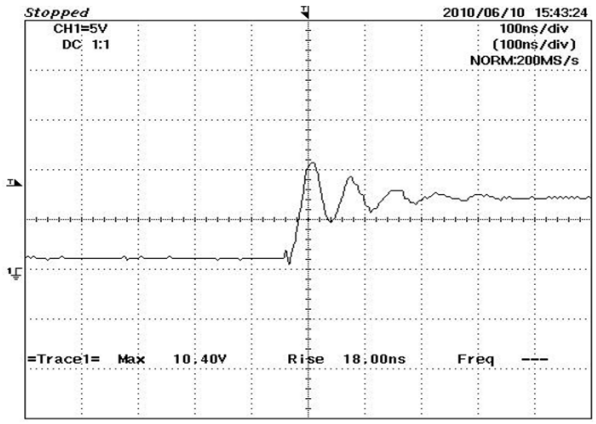

Figure 3. Voltage-time diagram across the plasma reactor

D. Measuring system

Pulse parameters were measured by using a voltage divider of ratio 2000:1 (EP-50 K, PEEC) and the digital storage oscilloscope (DL 1540: 8 bits 200 MS/s, 150 MHz, Yokogawa). The consumed power was measured from the input side by two reactor principle using a digital wattmeter. To measure power in this method two identical reactors are used. The power consumed by a plasma reactor is the difference of the power given to two identical plasma reactors (say w1) and that given to single plasma reactor (say w2) assuming constant power consumption in the transformer and RSG. The power given to reactor is mathematically denoted by w = (w1"w2). This input power can be expressed in J/L as follows:

$$\text{Specific Energy Density}(J/L) = \frac{\text{Discharge Power (watt)}}{\text{Gas Flow Rate (l/sec)}}$$

E. Plasma Reactor

The plasma reactor is one in which the high voltage pulse from the RSG is applied to produce the much needed electrical discharge to initiate the reaction to remove NO and NO_X emissions from the diesel exhaust. In general, the plasma reactor consists of a stainless steel electrode placed inside a glass tube/acrylic resin. The reactor here used is cross flow reactor (CFR). The CFR is of nine tubular reactors in parallel connected with electrodes. The dimensions of the CFR is 350mm x 250 mm x 20mm. The outer diameter of the CFR is 20mm and the inner diameter is 18mm. The reactor has one inlet and one outlet.

F. Flue Gas Analyzer

The gas after the treatment in the plasma reactor is let into the flue gas analyzer to analyze the proportions of the gas components. For each flow rate the concentration of the exhaust is recorded. The analyzer records the concentration of various gases like NO_X, NO, CO, CO_2, and O_2 by a set of infra-red sensors. The concentrations of the gas components are noted by applying various magnitude of voltage and the concentrations of all the relevant gases are recorded. The analyzer has a pump that is capable of taking the exhaust gas at 2 l/min from the plasma reactor.

III. Results and Discussion

Here, we discuss the results of the experiments carried out at laboratory level using the setup described in Fig. 1. The experiments have been conducted at flow rates of 2, 5 and 10 l/min at no load conditions. During the experiment the pulse repetitive rate was 75 Hz and rise time 18-24nsec. The pulse voltages were applied in the range of 0-25 kV. The input power to the reactor was measured by using two reactor methods and then expressed as specific energy density (SED) in J/L. The initial concentration of pollutants in diesel engine exhaust is given in Table I.

TABLE I
INITIAL CONCENTRATION OF DIESEL ENGINE EXHAUST

POLLUTANTS	DIESEL ENGINE AT No Load
NO	120 ppm
NO_2	10 ppm
NO_X	130 ppm
CO	455 ppm
CO_2	0.1 % vol
O_2	17.5% vol

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A. Analysis with cross-flow reactor

The diesel exhaust taken for observation is led to the Cross Flow Reactor (CFR) and concentration of pollutants at different SED levels and various gas flow rate are analyzed here. The analysis is carried out under a temperature of 25 ^{0}C and no load condition.The variation of NO and NO_2 with specific energy density is shown in Fig. 4. It is evident from the Fig. 4 that the concentration of NO steadily decreases with the increase of SED whereas NO_2 increases to certain value after that NO_2 also comes down. This signifies the production of radicals due to production of plasma and subsequent removal reactions taking place in the reactor chamber. Also, it can be observed that, on increasing gas flow rate, the SED, at which, a particular reduced concentration of NO_X/NO can be achieved, is reduced.

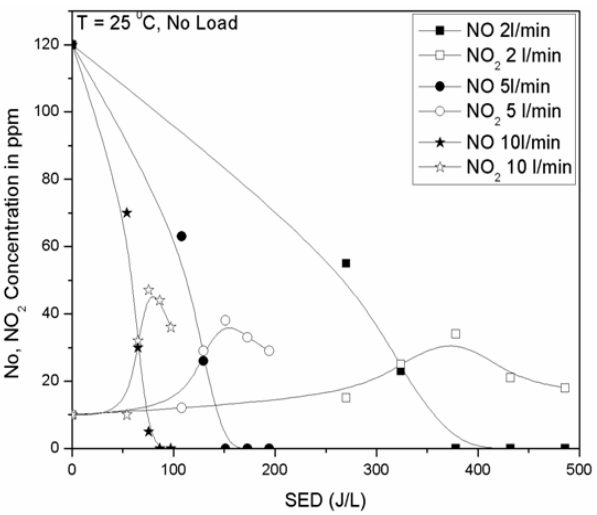

Figure 4. NO and NO_2 Concentration in ppm at different specific energy density

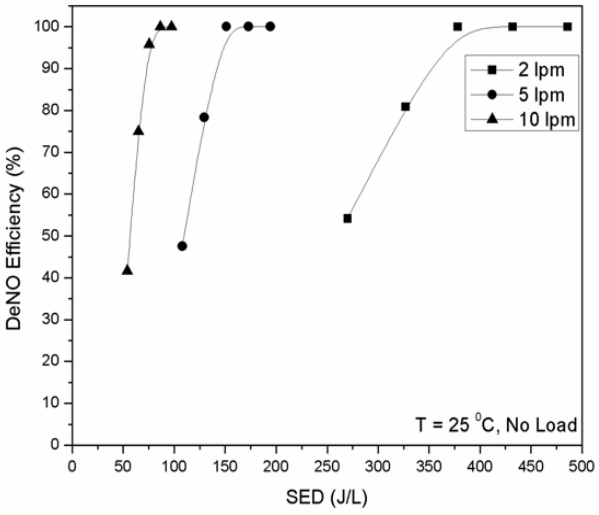

Figure 5. DeNO Efficiency at different specific energy density

NO and NO_X removal efficiency is shown is Fig. 5 and 6 respectively. The removal of NO_X is due to the oxidation of NO to NO_2 by O/OH radicals and as well as reduction of NO to N_2 by N radicals. In the filter exhaust plasma treatment, the O/OH radicals effectively help oxidation of NO to NO_2, thus leading to a high NO_X removal efficiency. The high electric field results in high energetic electrons. These electrons collide with other background gas molecules, resulting in production of more excited species and ions. However, most of them get quickly converted to radicals as given by following reactions.

The possible reaction pathways responsible for NO and NOx removal can be summarized as below [5-7]:

NO-NO_2 conversion reactions involving O/O_3/NO_3 radicals

$$O_2 + e \rightarrow O + O + e \qquad k=1.1\times10^{-12}\ cm^3/s \qquad (1)$$

$$NO + O \rightarrow NO_2 \qquad k=1.4\times10^{-12}\ cm^3/s \qquad (2)$$

$$O_2 + O \rightarrow O_3 \qquad k=1\times10^{-14}\ cm^3/s \qquad (3)$$

$$NO + O_3 \rightarrow NO_2 + O_2 \qquad k=1.8\times10^{-14}\ cm^3/s \qquad (4)$$

$$NO + NO_3 \rightarrow NO_2 + NO_2 \qquad k=1.5\times10^{-11}\ cm^3/s \qquad (5)$$

NO reduction reactions

$$N_2 + e \rightarrow N + N + e \qquad k=3.0\times10^{-16} \qquad (6)$$

$$NO + N \rightarrow N_2 + O \qquad k=2.1\times10^{-11} \qquad (7)$$

NO_2 conversion reaction

$$NO_2 + O \rightarrow NO + O_2 \qquad k=10^{-11}\ cm^3/s \qquad (8)$$

$$NO_2 + O_3 \rightarrow NO_3 + O_2 \qquad k=1.8\times10^{-17}\ cm^3/s \qquad (9)$$

$$NO_2 + NO_3 \rightarrow N_2O_5 \qquad k=1.8\times10^{-11}\ cm^3/s \qquad (10)$$

$$NO_2 + N \rightarrow N_2O + O \qquad k=3\times10^{-12}\ cm^3/s \qquad (11)$$

(Where k = rate constant)

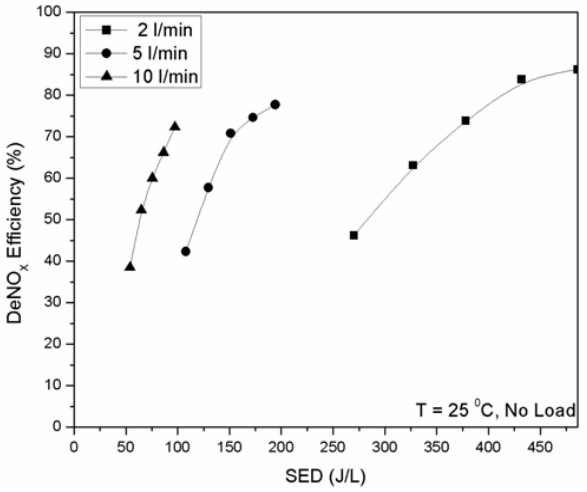

Figure 6. DeNOx Efficiency at different specific energy density

In Fig. 5, it is seen that with increase in SED value NO concentration decreases rapidly and after some value becomes zero. Here all of NO is not getting removed instead some of them gets converted to NO_2. At 97 J/L the NO removal efficiency achieved is 100 % with a gas flow rate of 10 l/min. It is evident from Fig. 6, that the NO_X removal efficiency is about 76% at 97 J/L with gas flow rate of 10 l/ min.

With industrial point of view it is very much important to know the energy needed for NO and NO_X molecule to be

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removed. The eV/NO and eV/NO_X can be calculated using the following formula:

$$\text{Energy Cost Per NO Molecule} = \frac{\text{power input to reactor} \times 6.25 \times 10^{18}\,\text{eV/sec}}{\text{NO Molecule Removed}}$$

Where

$$\text{NO Molecules Removed} = \eta_{DeNO} \times NO_{Initial}\,(\text{ppm}) \times 10^{-6} \times 2.45 \times 10^{19}\,(\text{molecules/cc}) \times \text{gas flow (cc/sec)}$$

$$\text{Energy Cost Per } NO_X \text{ Molecule} = \frac{\text{power input to reactor} \times 6.25 \times 10^{18}\,\text{eV/sec}}{NO_X \text{ Molecule Removed}}$$

Where

$$NO_X \text{ Molecules Removed} = \eta_{DeNO_X} \times NO_{Initial}\,(\text{ppm}) \times 10^{-6} \times 2.45 \times 10^{19}\,(\text{molecules/cc}) \times \text{gas flow (cc/sec)}$$

The eV/NO and eV/NO_X molecule at different gas flow rate is shown in Fig. 7 and 8. It is seen that with increase in gas flow rate the eV/NO and eV/NO_X decreases because eV required per molecule to be removed is indirectly proportional to the gas flow rate. From Fig. 8, it is evident that 206 eV is required for removal of one NO molecule at gas flow rate of 10 l/min, whereas 263 eV is required to remove one NO_X molecule. In Fig. 7, the eV required is increases when the input energy to the reactor is increased even after removing 100% of NO.

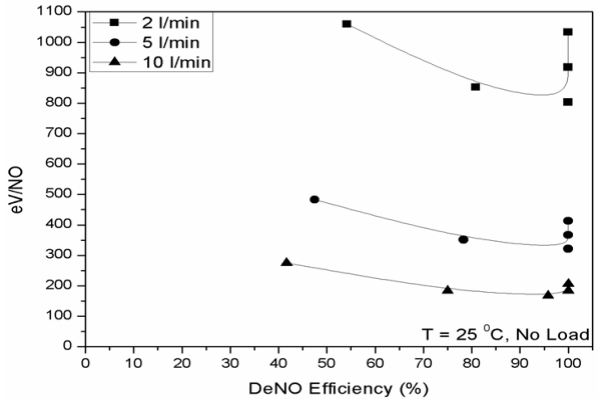

Figure 7. eV/NO molecule at different gas flow rate with respect to DeNO efficiency

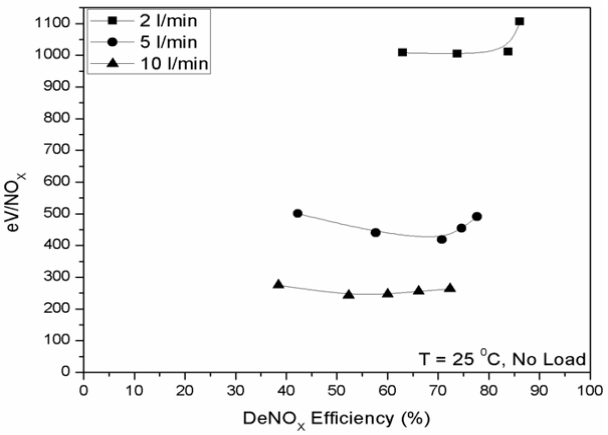

Figure 9. eV/NO_X molecule at different gas flow rate with respect to $DeNO_X$ efficiencyConclusion

The studies on the Cross Flow Reactor have been made under different flow rates (2, 5 and 10 l/min) and efficiency of removal is found to be reducing with the increase in the flow rate. The concentration of the gas molecules NO and NO_X are reducing with the rise in the applied potential in the chamber. The DeNO and $DeNO_X$ efficiency achieved with cross flow reactor is significant. The DeNO efficiency achieved at 97 J/L is 100% whereas NO_X removal efficiency was about 72% with gas flow rate of 10 l/min. A further study can be made with gas high gas flow rate. With about 263 eV one NO_X molecule can be removed from the exhaust gas.

At higher flow rate the Specific Energy Density (SED) is reducing due to the increase in the amount of molecules that pass through the reactor with the same energy density for an applied potential.

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